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CAPTURE CROSS SECTIONS FOR FAST NEUTRONS

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INTRODUCTION

Cross section measurements made with neutrons from chain-reacting piles have been mainly those made at or near thermal energy. Although piles have a high flux of neutrons extending all the way from fission energies to thermal energy, the great spread in energy has made cross section measurements at specific energies exceedingly difficult. It was the purpose of the present measurements to isolate a particular group of fast neutrons, the nascent fission neutrons of energy about 1 Mev, and use them for measurement of absorption cross sections. Although the fission neutrons themselves show a large spread in energy, the measured cross sections can be correctly interpreted because of the smooth variation of cross section with energy. The disadvantage of the energy spread is compensated by the large available flux, absolute calibration, and steadiness of the pile as a fast neutron source. The absorption cross section for thermal neutrons varies irregularly from isotope to isotope, depending on the proximity of the closest resonance level. As the neutron energy is raised, however, the width and density of the levels increase until, even with a monoenergetic neutron source, the observed cross section becomes an average over several levels. The absorption cross section at such high energies thus depends on the average behavior of the resonances, and is expected to be a slowly varying function of neutron energy and atomic weight.

The theory of resonance absorption of neutrons in terms of the "Breit-Wigner" formula has been treated by many authors. The discussion of Feshbach, Peaslee, and Weisskopf[1] is quite convenient for the present measurements. They show that the average absorption cross section for fast neutrons in a small energy band, ΔE at E, is given by

$$\sigma_a^{l} = \frac{\pi}{k^2}(2l+1)\ \frac{2\pi(\Gamma_{ln}\,\Gamma_{al})_{av}}{D_l\left[\,(\Gamma_{nl})_{av} + (\Gamma_{al})_{av}\,\right]} \qquad (1)$$

where σ_a^{l} is the contribution to the cross section of neutrons with angular momentum $l\hbar$ and wave number k, Γ_{nl} and Γ_{al} are the neutron and absorption widths for the many levels involved ("av" signifies average value), and D is the level spacing. It is seen that where $\Gamma_n > \Gamma_a$, as is true for fission neutrons, the variation of cross section with energy will be as $1/E$. In addition, any variation of cross section from isotope to isotope in this energy region would reflect the variation mainly in level spacing, D. Thus it would be expected that the cross section at 1 Mev would increase with atomic weight, rapidly at first (as D decreases with increasing complexity of the nucleus), and then should change only slowly for heavy nuclei (as D changes slowly - see reference 1, p. 155). Of course any unusual values of the neutron binding energy might be expected to give unusual values of D and hence of cross section. Measurements of absorption cross sections thus give information on the average level behavior, particularly the level spacing, D. It is because of this dependence of the cross section on the average level behavior that the spread of the fission distribution does not prove to be a serious complicating factor in the present measurements.

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* This is a more detailed account of work reported in MDDC - 27.

At the time the present experiments were begun, some experimental results were available on absorption cross sections for neutrons in the region of 1 Mev, based on work with cyclotrons, Van de Graaffs, Ra-α-Be, and photo-neutron sources. The reported cross sections were mainly for heavy nuclei (large cross sections) and in general were quite uncertain in regard to absolute calibration because of the difficulty of measuring fast neutron flux, although some of the recent Los Alamos results had good absolute calibration.

EXPERIMENTAL METHOD

Unmoderated fission neutrons were obtained (Figure 1) by irradiating a plate of uranium metal in a beam of slow neutrons from the thermal column of the Argonne heavy-water pile. A foil placed inside heavy cadmium and near such a plate receives only unmoderated fission neutrons if all slowing down materials are carefully excluded. As the capture cross section varies as 1/E, the presence of only a small number of moderated neutrons can vitiate the results. The flux of fast neutrons at the foil can be determined as accurately as the thermal flux can be measured, because the ratio of the fast fission flux to the thermal flux can be calculated from the geometry of the plate.

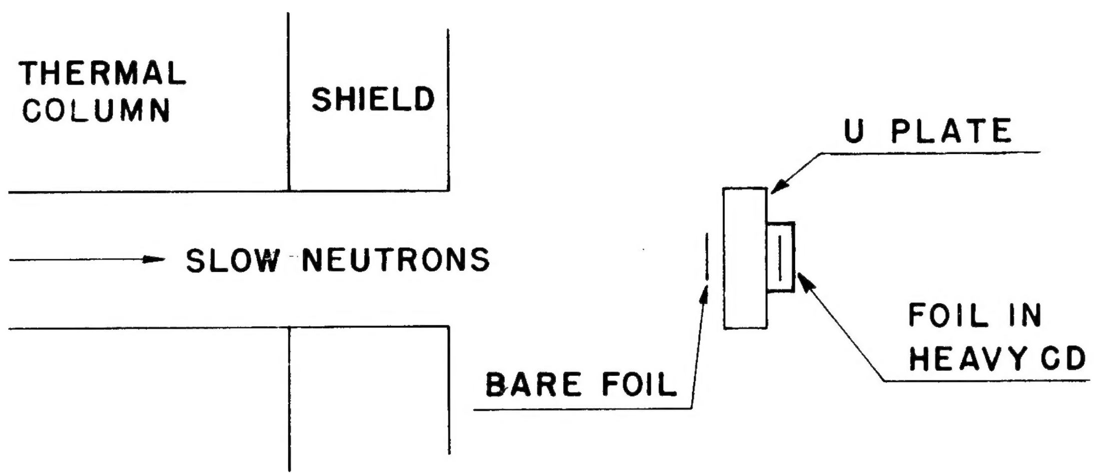

Figure 1.

The procedure, in brief, is to activate a cadmium covered foil of the material of interest with fission neutrons while an identical bare foil is simultaneously activated with thermal (plus fission) neutrons. The two foils are then counted on a GM counter. The fast activation cross section is obtained directly from the counting-rate ratio, the known thermal cross section for the particular activity detected, and the calculated ratio of fast to slow flux at the plate (which is approximately unity). In this way no corrections are necessary for self absorption in the foil, complicated decay schemes, or efficiency of the counter. In some cases where the thermal cross section was doubtful, it was determined from the bare-foil activation, taking into consideration the thermal flux and the counter efficiency. Because the fast cross section is often only one-thousandth of the thermal cross section, it was found extremely important to use a thick, tight cadmium cover for the fast irradiation to prevent significant leakage of thermal neutrons. A slight activation sometimes resulted from the small number of resonance neutrons present in the thermal column if the substance had a high resonance activation could be estimated easily however by running a cadmium-covered foil with the uranium plates removed. The resulting cross section is, of course, an average over the fission spectrum, but, because the variation of $\sigma(n,\gamma)$ with energy is about the same for different elements near 1 Mev

Table 1.

Isotope	Activity	Thermal σ_{act} (Isotopic)	Fast σ_{act} (Isotopic) = σ_a	Remarks
Na^{23}	18.8 hour	0.55 barns	0.29 mb	
Mg^{26}	10.2 min	0.049	0.60	
Al^{27}	2.4 min	0.23	0.40	
Cl^{37}	37 min	0.61	0.80	
K^{41}	12.4 hour	1.0	2.9	
V^{51}	3.9 min	5.5	2.2	
Co^{59}	10.7 min	0.73	.23 } 9.2	The total σ_a is based on the assumption that the isomeric ratio of 30/.72 holds for fast neutrons.
	5 year	30	-	
Mn^{55}	2.6 hour	11.5	3.5	
Ni^{64}	2.6 hour	3.4	6.7	See text
Cu^{63}	12.8 hour	3.1	8.9	
Cu^{65}	5 min	2.0	6.0	
Zn^{68}	57 min	0.89	7.1 } 20.1	Isomeric ratio: thermal 10.5; fast 0.55
	13.8 hour	0.085	13.0	
Ga^{69}	20 min	1.52	22.7	
Br^{79}	18 min	8.9	30 } 44	Isomeric ratio: thermal 3.0; fast 2.1
	4.4 hour	3.0	14	
Br^{81}	34 hour	2.3	17	
Rb^{87}	17.5 min	0.14	45	
Cb^{93}	6.6 min	1.4	52	See text
Mo^{98}	67 hour	0.39	32	
Mo^{100}	14.6 min	0.22	15	
Rh^{103}	44 sec	150	103 } 119.5	Isomeric ratio: thermal 11.7; fast 6.3
	4.2 min	12.8	16.5	
Ag^{107}	2.3 min	48	127	
Ag^{109}	22 sec	108	225	
In^{115}	13 sec	57	63 } 243	Isomeric ratio: thermal 0.36; fast 0.35
	54 min	157	180	
Sb^{121}	2.8 day	6.8	90	
I^{127}	25 min	6.8	107	
Ba^{138}	86 min	0.56	2.6	
W^{186}	23 hour	37	66	
Pt^{198}	31 min	3.6	59	
Au^{197}	2.7 day	96	121	
Hg^{204}	5.5 min	0.37	88	
Pb^{208}	3 hour	0.00046	1.8	
Bc^{209}	5 day	0.015	3.0	

(approximately 1/E), the measured cross sections can be correctly considered as capture cross sections at 1-Mev neutron energy (as calculated from the known fission spectrum and a 1/E energy dependence).

The technique was first checked by measuring Au^{197}, I^{127}, and In^{115}, which have large activation cross sections that have been measured as a function of energy. The results for these isotopes plotted at 1 Mev agreed well with the previous activation cross section values, as is shown in Figures 1, 2, and 3. From these first results it was concluded that the method gave the correct absolute capture cross sections at the calculated effective energy of 1 Mev. As the first elements had been done quite easily, the measurements were then extended to elements of lower atomic weight for which no cross sections were available. The method proved quite powerful in measuring the light elements because the high flux made activations possible for extremely low cross sections. A rough survey soon showed: (1) a rapid decrease of the activation cross section, σ_{act}, with decreasing atomic weight, A, for $A > 100$ (in this region σ varied nearly as A^4); (2) a rather smooth dependence of σ (isotopic) on A (the deviations from a smooth curve were not greater than the large experimental uncertainties associated with activation cross section measurements).

It was seen that if the smooth dependence of σ_{act} (isotopic) on A for $A < 100$ were true, then the total radiative capture cross section of an element for fast neutrons could be determined even if only one isotope of small abundance could be activated. This would be so because σ_a, total absorption, would be equal to σ_{act} (isotopic) for any isotope. The smooth dependence on A would also mean that the total absorption cross section of an element with no measurable activation could be predicted. Thus it would be expected that iron would have a cross section of 6.5 mb (mb = 10^{-3} barns) at 1 Mev and would increase about as 1/E for lower energies for at least several hundred kilovolts. The effect of structural materials and fission products in a "fast" pile could then be predicted, which is, of course, impossible for a thermal pile. Although the first measurements included only a few cross sections for heavy elements, it was obvious that the situation for $A > 100$ was not simple, as bismuth and lead showed much lower cross sections than the other heavy elements, all of which were in the 100 to 200 mb range.

After the early measurements, which were undertaken mainly to aid in fast pile design, a great many more cross sections were determined because the method proved to be useful from the standpoint of basic nuclear theory. The work was done at irregular intervals during the past several years, but only the final results, brought up-to-date, will be presented here.

DISCUSSION OF RESULTS

Table 1 contains the final cross section values for 31 isotopes, for 4 of which both activities of isomeric pairs were measured. For each isotope the activity studied, its thermal isotopic activation cross section, and the fast activation cross section (which, barring unknown isomers, will be equal to the atomic absorption cross section of the element, σ_a), are given. Figure 2 shows the cross sections plotted as a function of atomic weight and it is seen that the values for $A < 100$ follow a rapidly rising smooth curve within a factor of about 2. Thermal cross sections of course, do not behave in such a regular manner, and the curve of Figure 2 can actually be used to infer peculiarities in the thermal cross section if the fast cross section (based on the thermal) does not fall on the curve. Examples of this behavior will be given in the following discussion of individual isotopes.

Cb^{93}

At the time the present work was begun, it was thought that the thermal activation cross section for 6.6-min Cb^{94} was only 0.01 b while the total absorption (as measured by the effect on the pile) was known to be 1.4 b. The fast cross section measured in the present experiments, if based on a thermal value of 1.4 b, turned out to be 52 mb, which is on the smooth curve. This result implied that the total thermal activation cross section should be 1.4 b, and not 0.01 b (i.e. the difference was not caused by impurities as had been expected). The correctness of this procedure was shown when it was found by

Goldhaber and Sturm[2] that Cb^{94} decayed by isomeric transition and the 0.01-b value was low by just the efficiency factor for γ detection of a GM counter, the correct activation cross section being about 1.4 barns.

Co^{59}

With thermal neutrons, a strong 5-year and a 10.7-min isomer are produced, with cross section in the ratio 41 (this ratio may actually be lower because of uncertainity in the fraction of the 10.7-min activity which grows into the 5 year). In the present experiments, only the 10.7-min activity was measured, and the total fast activiation cross section was obtained by multiplying the 10.7-min isomeric cross section by 41. The agreement of the result, 9.2 mb, with the curve indicates that there is not a large shift in isomeric ratio with neutron energy in this case.

Ni^{64}

The early results of the present method, based on the extant thermal cross sections, gave a fast cross section that was far off the curve. It was then decided to remeasure the thermal cross section. The resulting atomic activation cross section for the 2.6-hour activity was 0.030 b as compared to the old 0.017 b (Seren's[3] value). In the meantime it was shown by Boyd and Goldhaber, that the 2.6-hour activity was Ni^{64} (0.88%) and not Ni^{62} (3.88%). The combined effect of these two corrections was to raise the fast isotopic cross section by a factor of about 7 and to bring it into agreement with the smooth curve.

Zn^{68}

Measurements of the isomeric activities of Zn^{68} (57 min and 13.8 hour) again gave values off the curve when the listed values[3] of the thermal cross section were used. The thermal measurements were then repeated with the result that the reported 1.09 b isotopic value for the 57-min activity was lowered to 0.89 b, and for the 13.8-hour activity the isotropic value was lowered from 0.31 b to 0.085 b. The new thermal cross sections led to a fast total activiation of 20.1 mb again, in agreement with the curve.

For the four isotopes for which both members of isomeric pairs were measured, the isomeric ratios (ratios of activation cross sections for the isomers) for thermal and fast neutrons are also given in Table 1. In the cases of In^{115}, Rh^{103}, and Br^{79} the ratio is the same, within experimental error, for fast and for slow activation. For Zn^{68}, however, the ratio changes from a thermal value of 10.5 to 0.55 for fast neutrons. The fast isomeric ratio is of course the average of the ratio over many resonance levels of the compound nucleus and might be expected to be different (and closer to unity) than the thermal ratio, which usually refers to a single level. The present results although rough are not inconsistent with this view since the changes which do show are in the correct direction. The behavior of Co^{59}, however, as discussed above, seems to indicate a large isomeric ratio which does not change greatly with neutron energy.

Among the heavy isotopes there are several marked exceptions to the general rule that the cross sections are roughly constant, namely, Ba^{138}, Pb^{208}, and Bi^{209}. Feshbach, Peaslee, and Weisskopf had already pointed out that lead and bismuth, because of their low absorption, must have an unusually large level spacing D, which would mean an unusually low neutron binding energy. However, as the same behavior in Ba^{138} was unexpected, attempts were made to find something wrong in the measurement or interpretation. Weisskopf suggested that perhaps competition by inelastic scattering could cause the low cross section in Ba^{138}. This possibility was checked by measuring the neutron capture cross section with Na-Be (900 kev) and Na-D_2O (230 kev) photo-neutrons. The cross section was unusually low, however, even at 230 kev where no inelastic scattering would be present. In the meantime, however, Mayer had shown that certain numbers of neutrons in nuclei formed stable "shells," and that the binding energy for the next neutron should be extremely low. The low cross section for Ba^{138} was then easily explained because it contains 82 neutrons, forming a closed shell (Pb^{208} and Bi^{209} each contain 126 neutrons, also a closed shell).

COMPARISON WITH OTHER MEASUREMENTS

As already discussed, the results for Au, I, and In were compared with the already existing data on cross section vs energy, as a check on the general method. After the other measurements had been made, an exhaustive survey by Way and the isotope information group at Clinton showed that there were quite a number of old results on capture cross section for fast neutrons.

In many cases the energies were not well-defined and the absolute cross section values were not good. However, if a set of values of a given experimenter is arbitrarily adjusted to bring his value for a standard substance, say Au^{197}, into agreement with Figure 2, then a comparison with the present values is possible.

Curves based on all the available data for a number of isotopes are given in Figures 3-17. All those isotopes are shown for which more than 3 points were available above 0.1 Mev. The sources of data, as marked on the figures, are as follows:

Dementi, V. S., and D. V. Timoshuk, Absorption of Rn + Be Neutrons, Comptes Rendus de l' Acadamie des Sciences de l' URSS, 27, 926.

Demers, P., H. H. Halban, R. Allen, and G. R. Bishop, Comparison of Cross Sections for the Capture of 220 and 900 kev Neutrons, Nature, 161, 727.

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Griffiths, J. H. E., The Absorption of Neutrons of Medium Energy, Proceedings of the Royal Society, A, 170, 513.

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Hughes - Present results.

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LA-1. Fischer, M. B., S. L. Friedman, A. O. Hanson, C. D. Klema, J. H. Williams, G. A. Linenberger, M. L. Perlman, E. Segre', and M. Deutsch.

LA-2. Linenberger, G. A., J. A. Miskel, J. M. Blair, M. Deutsch, K. I. Greisen, A. O. Hanson, R. F. Taschek, C. M. Turner, and J. H. Williams.

Recently additional measurements of activation cross sections have been made for those nuclei having 82 neutrons. In addition to Ba^{138}, already reported in the present paper, Pr^{141}, La^{139}, and Xe^{136} have been measured. The results for these isotopes are as follows:

Pr^{141}	11 mb
La^{139}	6 mb
Ba^{138}	2 mb
Xe^{136}	1 mb

These cross sections are definitely less because of nearby isotopes not containing 82 neutrons, which have cross sections of about 100 mb. The low cross sections of the 82 neutron isotopes thus furnish strong support for the hypothesis that 82 neutrons constitute a stable shell. It is interesting that isotopes show a regular increase of cross section with atomic number, which is to be interpreted as a decrease in stability as protons are added.

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2. Goldhaber, M., and W. Sturm, Phys. Rev., 70, 111 (1946).

3. Seren, L., H. Friedlander, and S. Turkel, Phys. Rev., 72, 888 (1947).

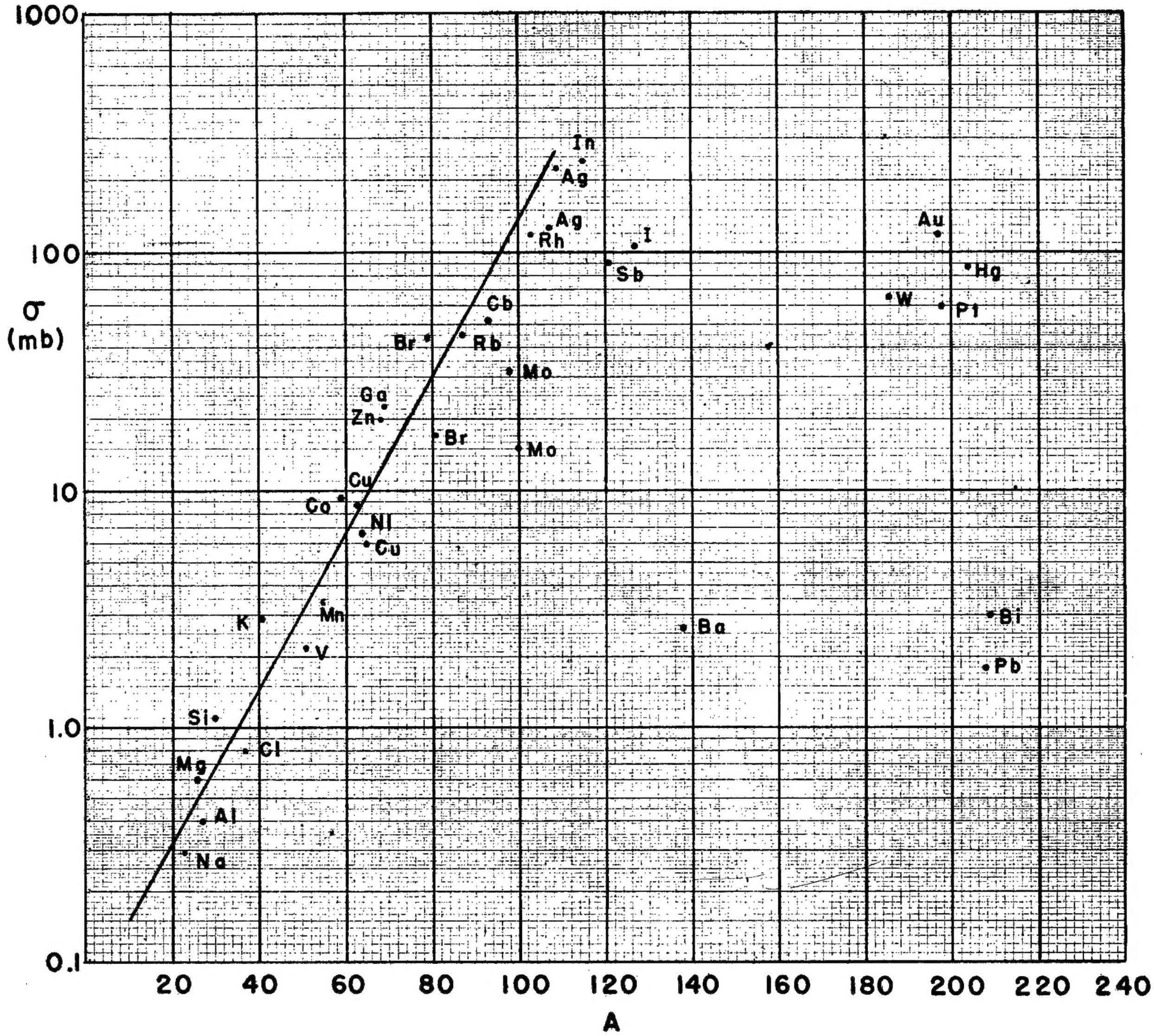


Figure 2. Isotopic activation cross sections for fission neutrons.

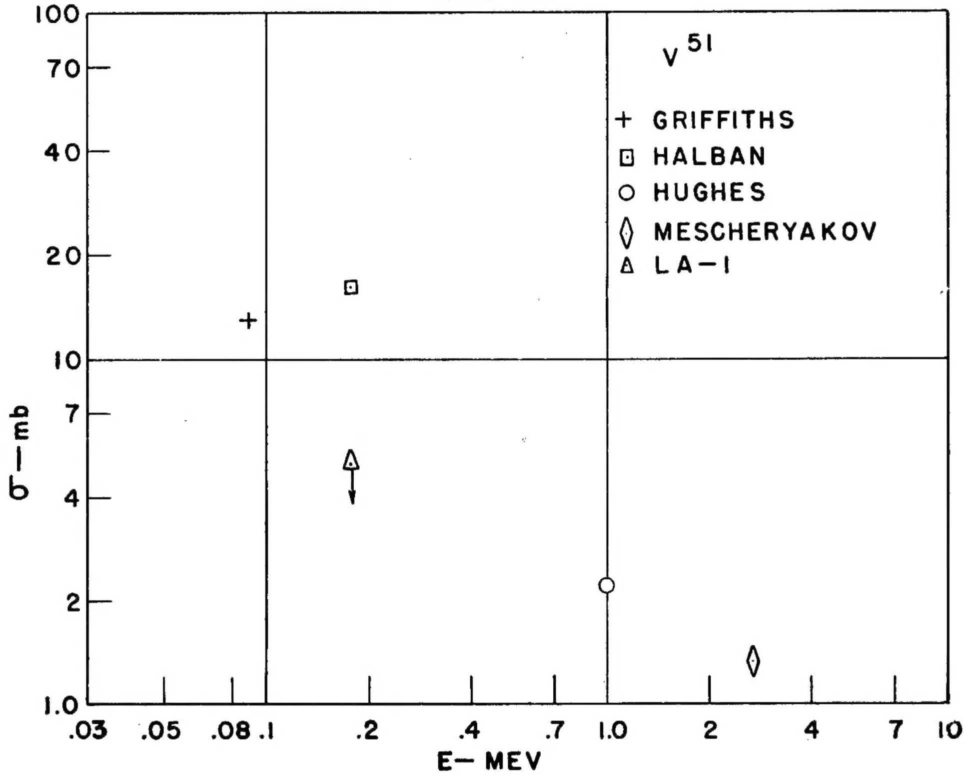
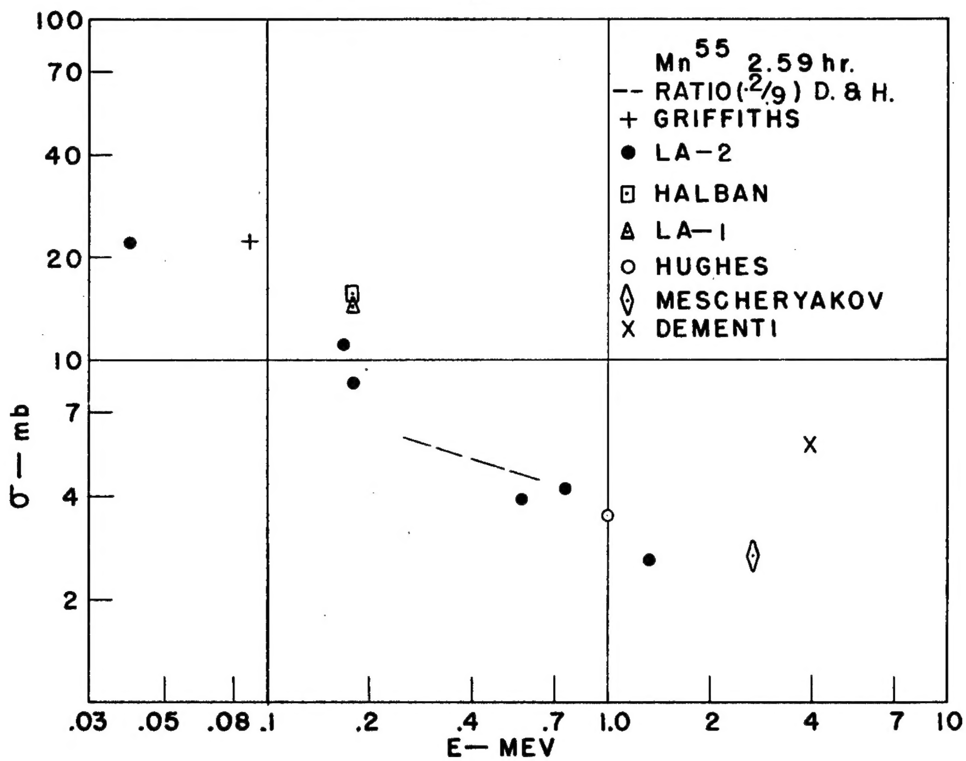


Figure 3.



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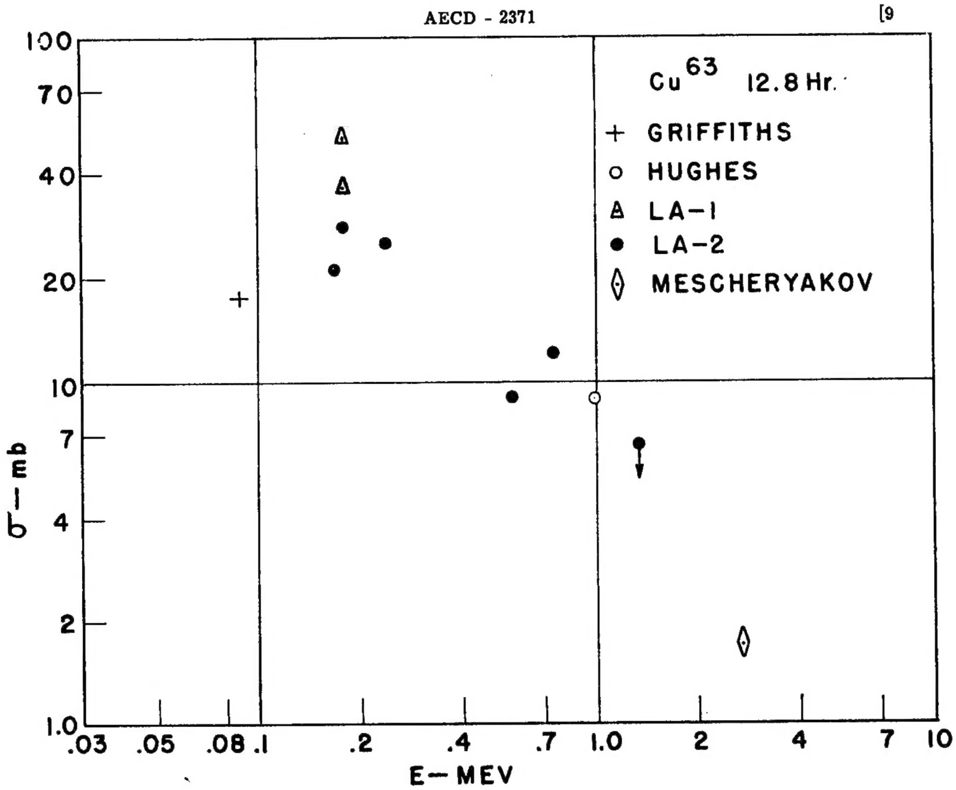

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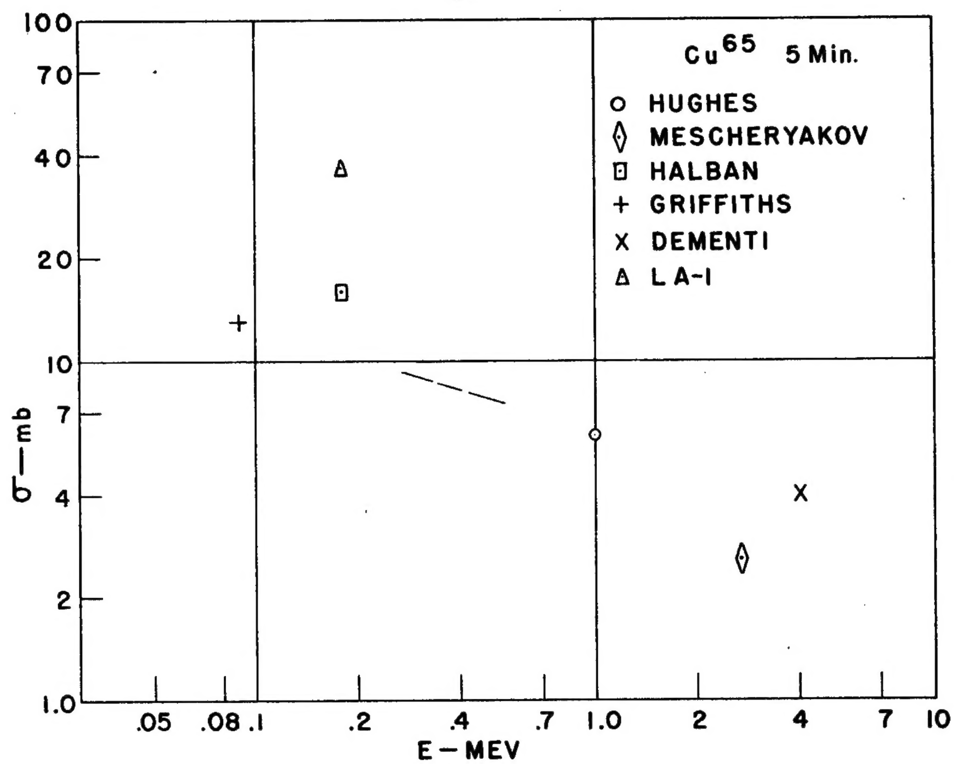

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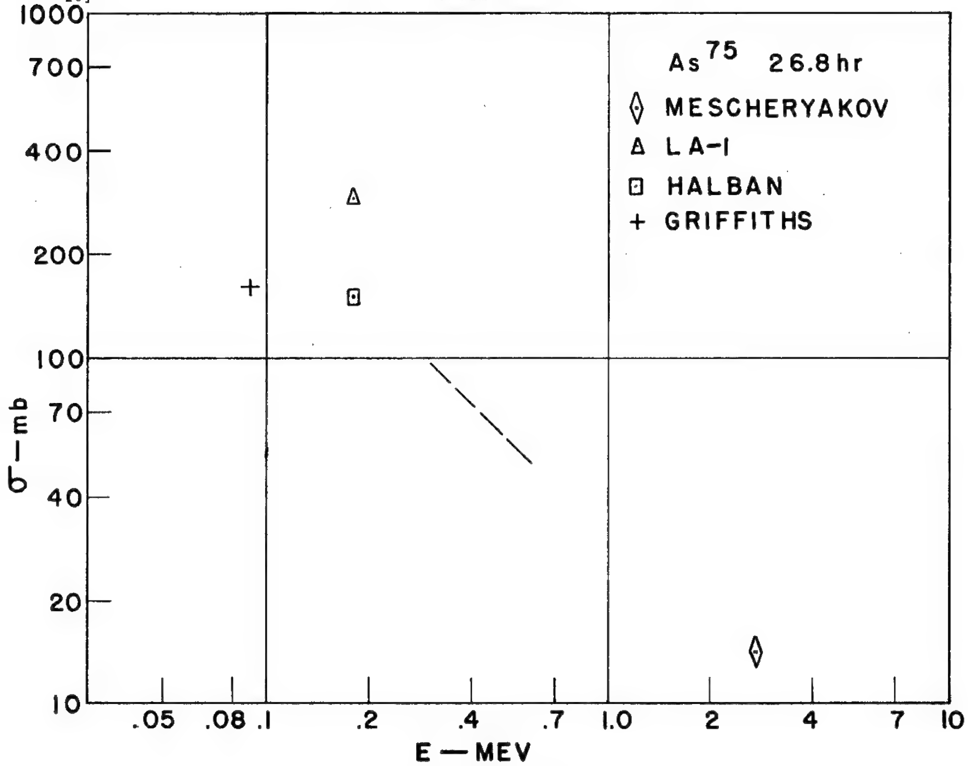

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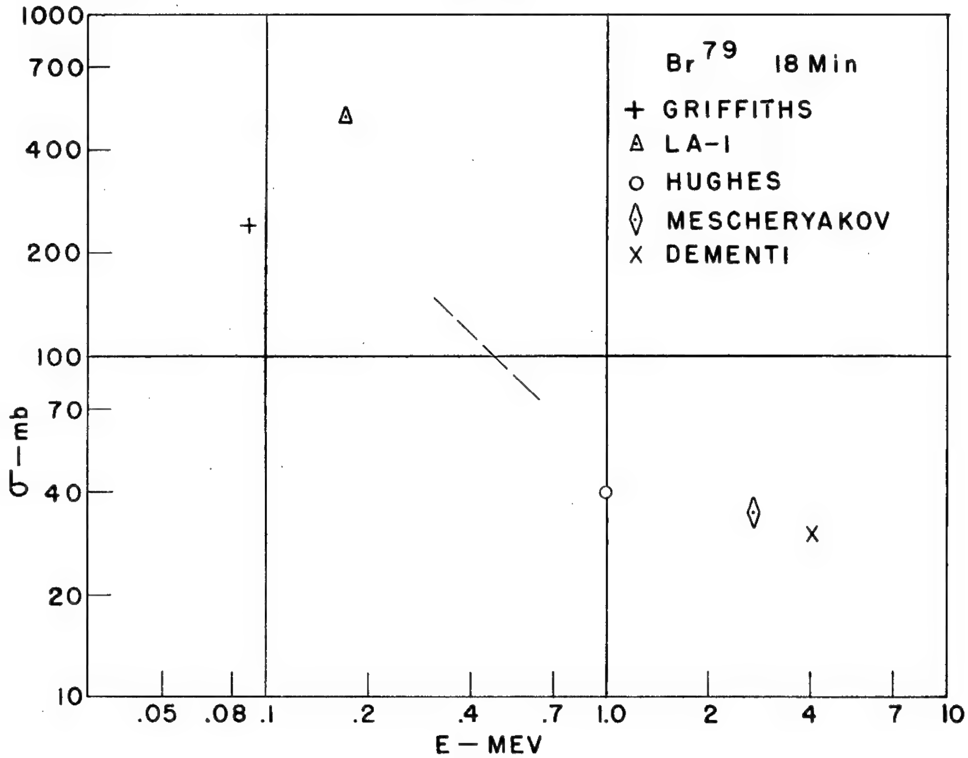

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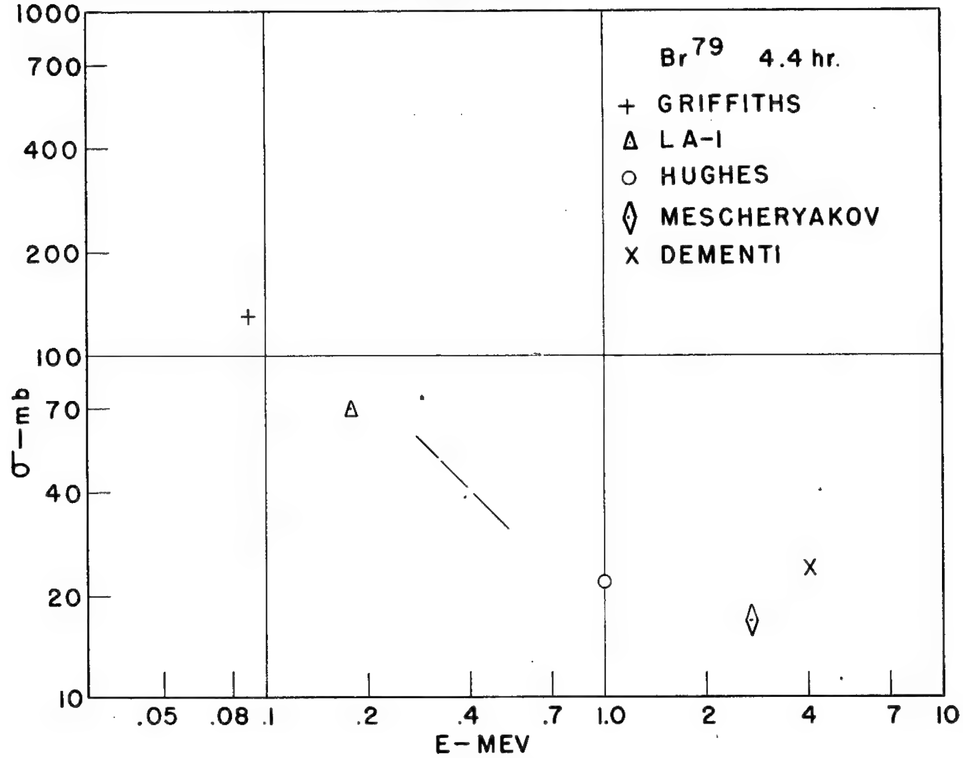

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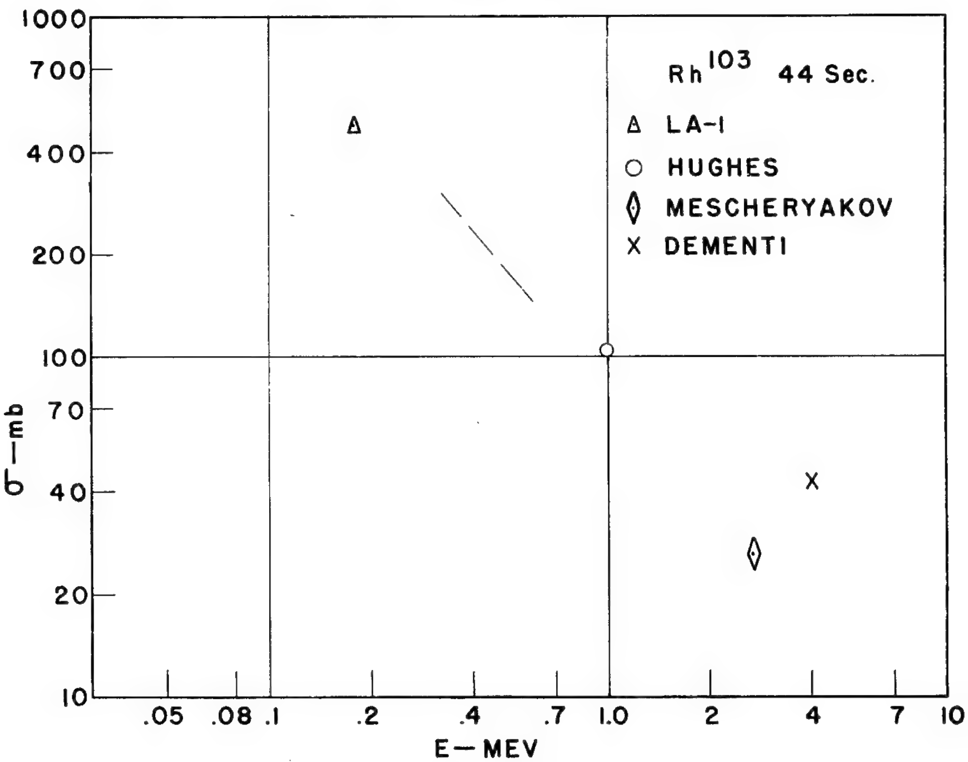

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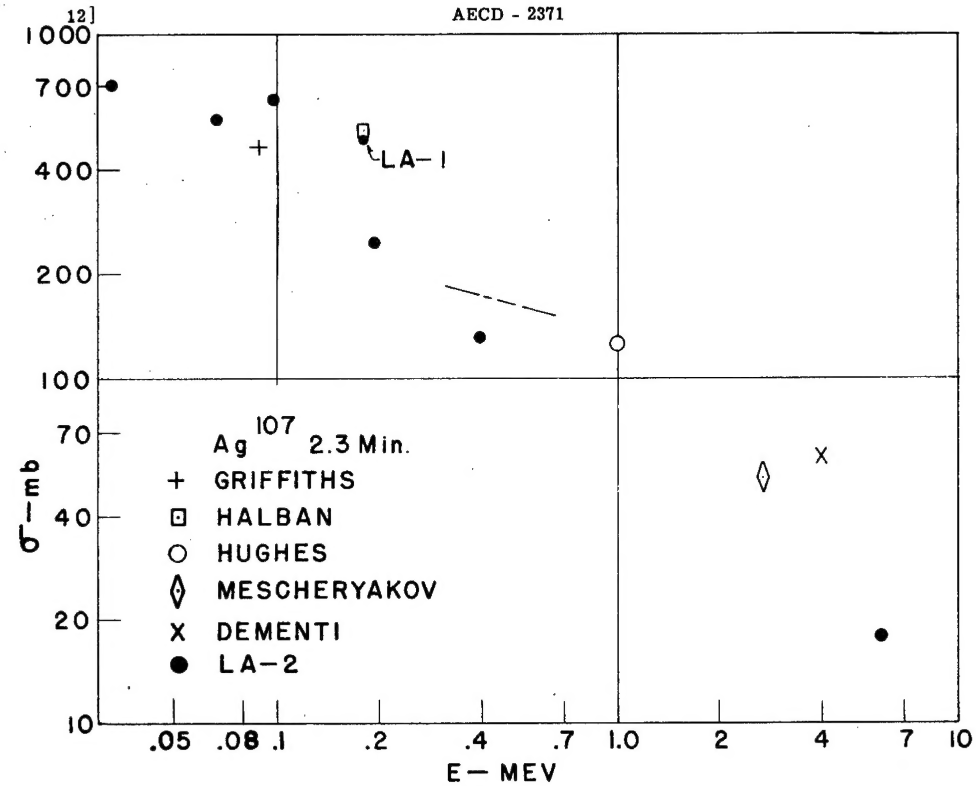
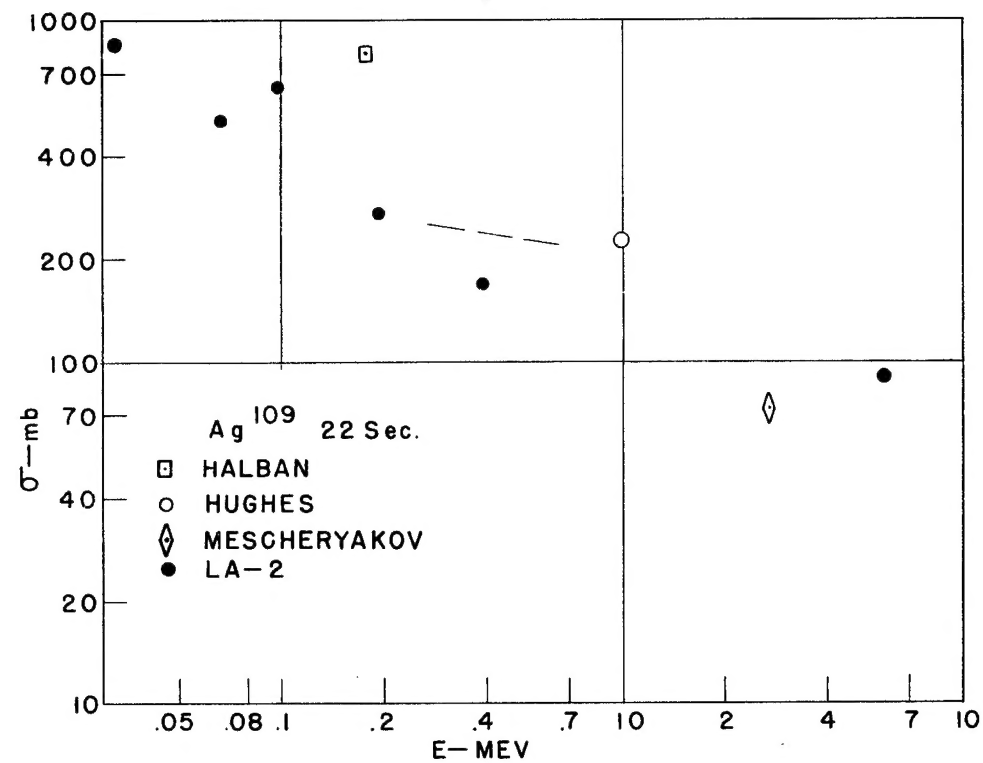


Figure 11.



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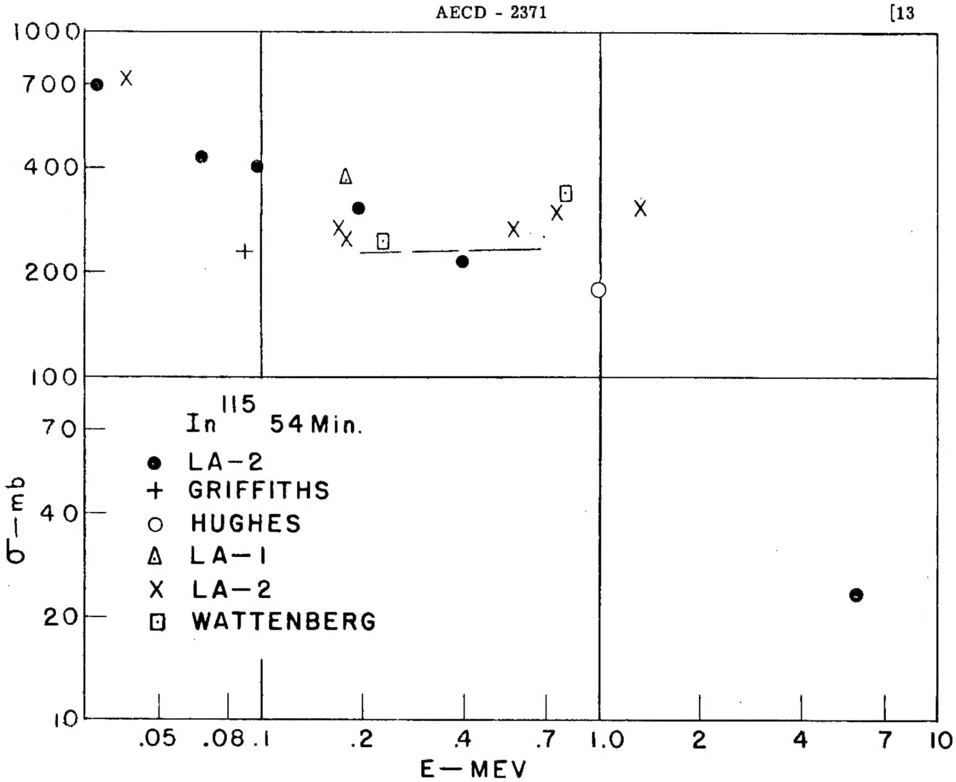

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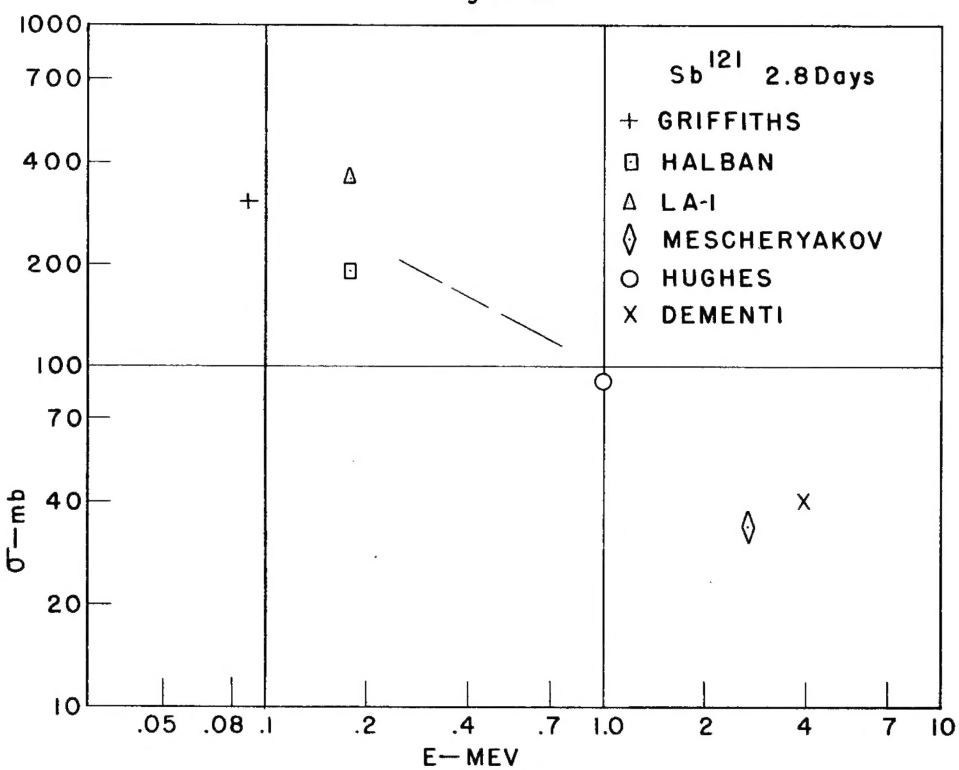

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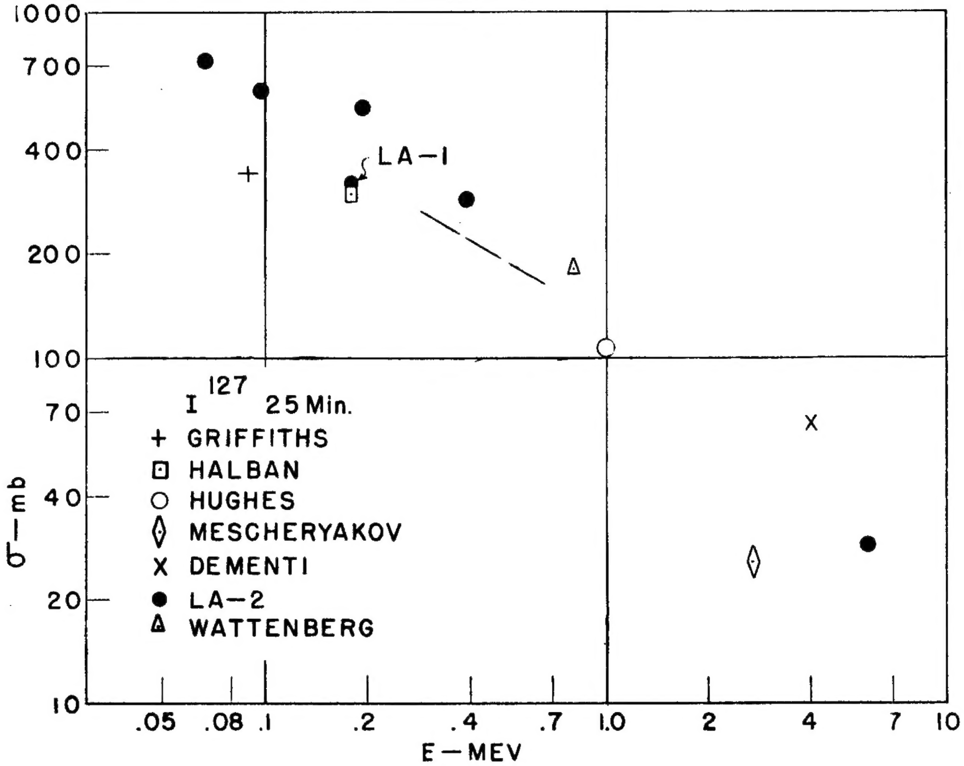

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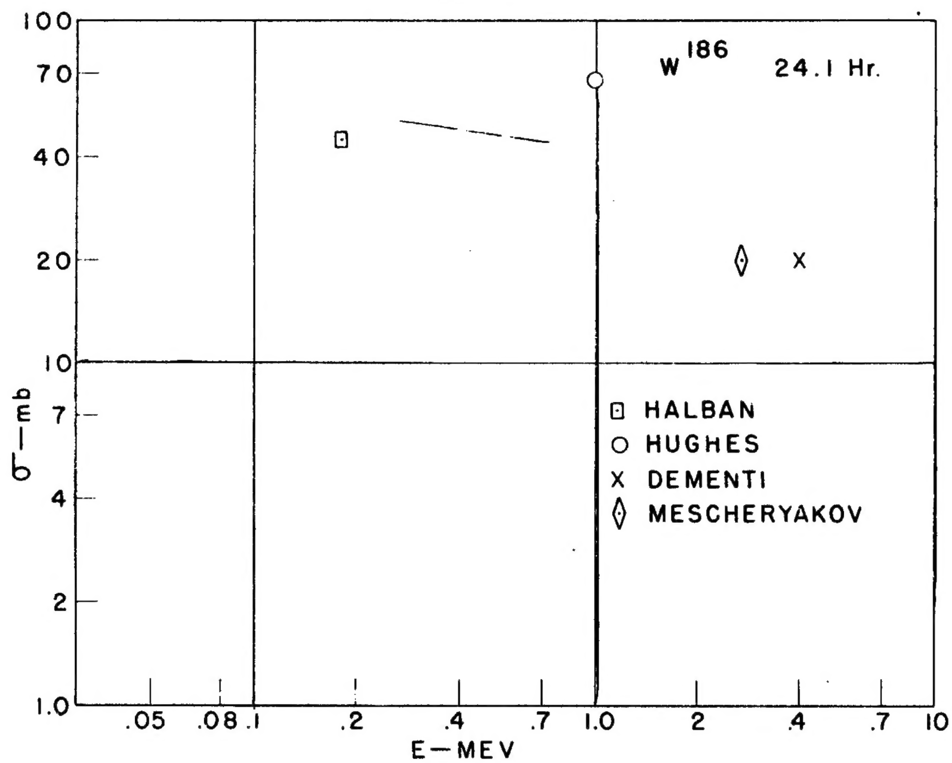

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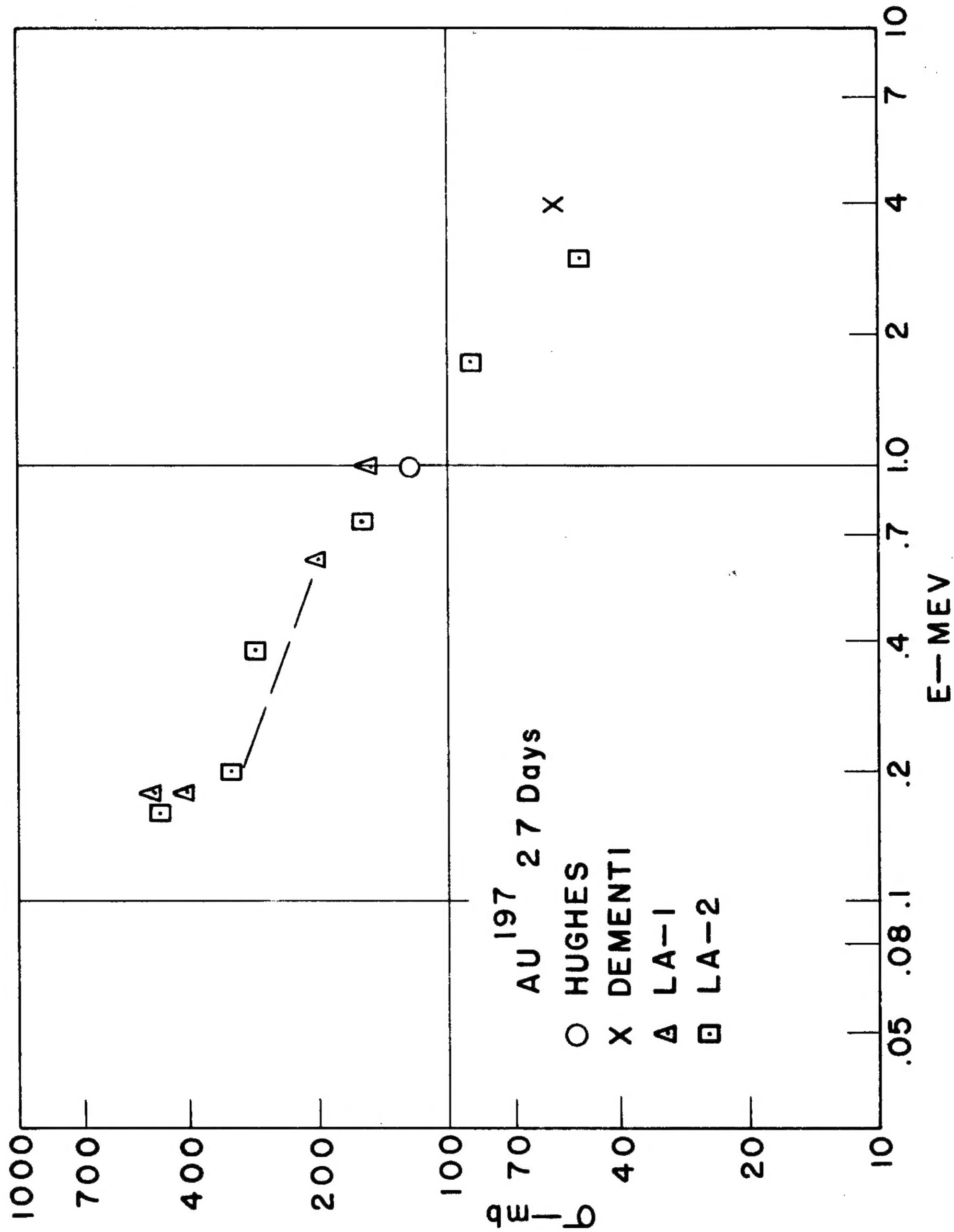

Figure 17.

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